Molecular Kinetic Computation of Turbulence in a Reacting Gas Flow

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Introduction

We consider the problem of computing chemically reacting gas flow. This problem is known to be not easy to cope with because of the fine structure of chemically reacting zone, where the reacting process is essentially at molecular level. Moreover it is combined with the complexity of turbulent flow field for the present problem. This situation makes it inevitably difficult for any approach based on continuum mechanics model. On the other hand, the approach such as molecular dynamics (MD) is certainly an orthodox way to this problem but unfortunately it is still far to be practical to the present case of chemical reaction combined with turbulence. Here we use as a second choice the molecular kinetic approach by the Boltzmann equation. There have been some studies of reacting gas flow along with this line but seldom of turbulent flow. The advantage of using molecular kinetic approach over continuum model to problems of computing compressible turbulence has been recognized (Bird 1997: Chen et.al 2003). Especially, the use of integral form of the Boltzmann equation based on its integral along the characteristic line is found to be effective to these problems (Sakurai and Takayama 2003), and it is expected to be so to the present case of turbulence combined with chemical reaction. Here we make an attempt to see its feasibility to this problem by considering the simplest case of turbulent flow for a binary gas mixture of oxygen atoms and molecules at a reasonably high temperature above 2,000 K with dissociation and recombination reactions in a square region bounded by constant temperature wall. For the sake of simplicity, we employ a co-planar model for the molecular kinetics and assume the BGK model for elastic collision term of the model equation. With reference to the discrete Boltzmann equations with reaction (Monaco and Preziosi 1991), we introduce a simple reaction term to the collision term. We set a random initial condition given by a local Maxwellian distribution function with mean velocity of random phase and magnitude to produce a turbulent flow, and see the development of turbulent oxygen gas mixture accompanying the reaction processes in a high temperature field.

Kinetic Model Equation and Computational Scheme

The kinetic model equation with chemical reaction is assumed here as

$$\mathrm{D}f_j / \mathrm{D}t = \nu_j (f_{je} - f_j) + (\mathrm{d}\,n_j / \mathrm{d}\,t)_c f_{je}^*, \qquad j = 1, 2 \qquad (1)$$

where f_j is the velocity distribution function of O ($j = 1$) or O_2 ($j = 2$) gas, f_{je} ($= n_j f_{je}^*$) is the Maxwellian distribution function based on the mean velocity $\boldsymbol{u}$ and the mean temperature T for gas mixture, ν_j is the elastic collision frequency, and ($\mathrm{d}\,n_j / \mathrm{d}\,t)_c$ is the production rate of the number density n_j due to the forward and reverse chemical reactions $O_2 + M \rightarrow 2\,O + M$ and $2\,O + M \rightarrow O_2 + M$, where the third body M denotes O or O_2. We assume that the distribution functions of reacting chemical species (reactants and products) are the Maxwellians based on the mean macroscopic properties for the mixed gas. This assumption can be considered to be adequate enough for reacting gases near thermal equilibrium, but we dare to apply this model to the present problem including nonequilibrium situations. We assume that the reactive collision frequency is much smaller than the elastic one. We assume that atom or molecule has only translation energy without rotation, vibration, and chemical energy except evaluating the production rate for chemical reaction.

The macroscopic properties n_j, n, ρ, $\boldsymbol{u}$ (u, v), and T for the co-planar gas are expressed in dimensionless forms as $n_j = \int f_j\,\mathrm{d}\,\boldsymbol{c}\, (= \int \int f_j\,\mathrm{d}\,c_x\,\mathrm{d}\,c_y)$, the total number density: $n = n_1 + n_2$, the mass density: $\rho = n_1 + 2\,n_2$, $\rho\,\boldsymbol{u} = \int \boldsymbol{c}\,(f_1 + 2 f_2)\,\mathrm{d}\,\boldsymbol{c}$, and $n\,T = \int C^2\,(f_1 + 2 f_2)\,\mathrm{d}\,\boldsymbol{c}$, where $\boldsymbol{C} = \boldsymbol{c} - \boldsymbol{u}$. As normalizing quantities, we use the side length L of the square wall, the gas temperature T_{00}, the atom number density n_{100}, and the most probable speed of atoms $c_{00} = (2\,k\,T_{00} / m_1)^{0.5}$ at a reference state, for which we assume a fully dissociated condition ($n_{10} = 1$, $n_{20} = 0$) at $T_{00} = 5{,}000$ K. The normalized Maxwellian distribution function is expressed as $f_{je} = j\,n_j\,(\pi\,T)^{-1} \exp(-j\,C^2 / T)$. We use the elastic collision frequency for hard-sphere molecules as

$$\nu_1 = (2T/\pi)^{0.5}\,\{2^{0.5}\,n_1 + (1.5)^{2.5}\,n_2\}/K_n, \quad \nu_2 = (2T/\pi)^{0.5}\,\{(1.5)^{2.5}\,n_1 + 4\,n_2\}/K_n.$$

The Knudsen number is defined by $K_n = \lambda_{00} / L$, where λ_{00} is the reference mean free path for oxygen atoms. We use the species production rate on the basis of the Lighthill's ideal dissociating gas model (Lighthill 1957: Vincenti and Kruger 1965), which can be described in a dimensionless form for oxygen atom as

$$\mathrm{d}\,n_1 / \mathrm{d}\,t = C_1\,T^{-S}\,\rho^2\,\{\alpha + 0.1\,(1 - \alpha)\}\,\{(1 - \alpha)\,\exp(-T_d / T) - (\rho/\rho_d)\,\alpha^2\}/K_n,$$

where α is the mass fraction of oxygen atom defined by $\alpha = n_1 / \rho$, T_d is the normalized characteristic temperature for dissociation as $T_d = T_d^* / T_{00} = 11.9$ ($T_d^* = 59{,}500$ K), and ρ_d is the normalized characteristic density for dissociation as $\rho_d = \rho_d^* / \rho_{00}$ ($\rho_d^* = 150$ g cm^{-3}). Using the reaction rate constants of O_2 dissociation and recombination (Park 1995), we obtain $C_1 = 6.26 \times 10^4$ and $S = 1.5$. For oxygen molecule, we use the relation $\mathrm{d}\,n_2 / \mathrm{d}\,t = -0.5\,\mathrm{d}\,n_1 / \mathrm{d}\,t$,

which is derived from the law of mass conservation.

In our computational scheme, we use the integral form of the model equation (1) along the characteristic line to avoid the difficulty of finite difference schemes for the expected complex structure of a turbulent flow field. It is expressed for a small time interval Δt as

$$f_j(c, x + c \Delta t, t + \Delta t) - f_j(c, x, t) = \Delta t \,(\text{collision terms}),$$

and can be rewritten by making the transformation $x + c \Delta t \rightarrow x$ to give

$$f_j(c, x, t + \Delta t) = f_j(c, x - c \Delta t, t) + \Delta t \,(\text{collision terms}). \tag{2}$$

This equation is a basis of our computational scheme. The physical and velocity spaces are discretized into square cells or grid points, and the computational procedure is divided into two phases in each time step. In the first translational phase, we construct $f_j^{(1)}(c, x, t + \Delta t)$ from $f_j^{(0)}(c, x - c \Delta t, t)$ without the collision terms. The value of $f_j^{(0)}$ at $x - c \Delta t$ is estimated from a linear interpolation among its grid-point values, since the position $x - c \Delta t$ is not necessarily on a grid point. In the second collision phase, we modify $f_j^{(1)}$ by using a formula

$$f_j^{(2)}(c, x, t + \Delta t) = f_j^{(1)} + \Delta t \,\{ \nu_j (f_{je} - f_j) + (d\,n_j / d\,t) f_{je}^* \}^{(1)}. \tag{3}$$

We assume the diffuse reflection at a non-catalytic wall as a boundary condition for the square wall surrounding the hot gas region. Initially, we select the gas temperature T_0, the wall temperature T_{w0}, and the number densities n_{10} and n_{20} at a uniform state over the whole region. Then, we consider a turbulent reacting gas flow starting from a random initial condition at $t = 0$. Here we set local Maxwellian distribution functions for O and O_2 gases with common flow velocity u_0, where the magnitude of u_0 obeys the isotropic energy spectrum $E(k) = K^2 k^3 \exp\{-2(k/k_0)^2\}$, $k_0 = 10$, $k = |k|$ for wave vector k, and K for an adjusting constant for the normalization, while its phase is given randomly as follows. Let $k = 2\pi b$, $b = (b_1, b_2)$, $b_i = 0, \mp 1, \mp 2, \ldots$ for $i = 1, 2$ and set $u_0 = (u_0, v_0)$ with $u_0 = \Sigma a_u(b_1, b_2) \sin 2\pi \{ b_1 x + b_2 y + \varepsilon_u(b) \}$, $a_u = (E / k^2)^{0.5} = k \exp\{-(k/k_0)^2\}$, and a similar expression for v_0, where $\varepsilon_u(b)$ are random numbers in $[0, 1]$ for each given combination of $b = (b_1, b_2)$ and $x = (x, y)$.

Results and Discussion

The computations are performed for several combinations of physical properties to be given as initial conditions, while we fix the characteristic density for dissociation as $\rho_d = 4 \times 10^7$, for which active oxygen dissociation and recombination reactions are expected in the temperature range around 2,500 K to 4,000 K, corresponding to the resultant chemical equilibrium mass fraction $\alpha_e = 0.04$ to 0.97. The resultant planar distributions of velocity, temperature, and atom mass fraction are shown in the following figures for an example case of $T_0 = T_w = 0.6$ (3,000 K), $n_{10} = 0.5$, and $n_{20} = 0.25$ ($\alpha_0 = 0.5$, $\rho_0 = 1$), where T_w is fixed throughout the calculation. The Knudsen number is given as $K_n = 0.005$, whose value is selected as an appropriate value

for turbulence calculation to be carried out by molecular models (Bird 1997). We use the maximum velocity value to get the initial Mach number $M_0 = 1.7$ and the corresponding Reynolds number $Re_0 = 500$.

The normalized computational range of x is 1 x 1 square, and is discretized into 100 x 100 square cells, which means the half side length of a cell corresponds to the reference mean free path. The normalized computational ranges of c_x and c_y are $-6 < c_x < 6$ and $-6 < c_y < 6$, and are discretized into 20 x 20 square cells. The time step Δt is selected as 0.0008 by considering the flight path length in a cell for the maximum molecular speed within the above molecular velocity range.

Figures 1 to 3 depict color contour maps, respectively, for the magnitude of velocity $|u|$, the temperature T, and the mass fraction of atoms α at $t = 0.024$. In each figure, we can see a fine structure of turbulence with several yellow spots, which mean hot regions for the temperature contour and fully dissociated regions for the mass fraction contour of atoms. The magnitude ranges of T and α are, respectively, 0.60 (3,000 K) $< T <$ 0.93 (4,650 K) and 0.45 $< \alpha <$ 0.85. This result indicates that the gas mixture is heated due to energy dissipation of turbulence to produce atoms by dissociation reaction within the high-temperature flow field. The cooler wall acts as a heat sink accompanying a gradual temperature decrease of the hot gas mixture to produce molecules by recombination reaction in a decaying process of turbulence.

Conclusion

The present kinetic model of the Boltzmann equation including a simple reaction term seems to work for analyzing the turbulent flow field accompanying chemical reactions. Especially, the results above suggest that it is useful for studying the coupling effect between chemical reactions and turbulence. Some advanced study will be required in future for the kinetic model to include the chemical energy.

References

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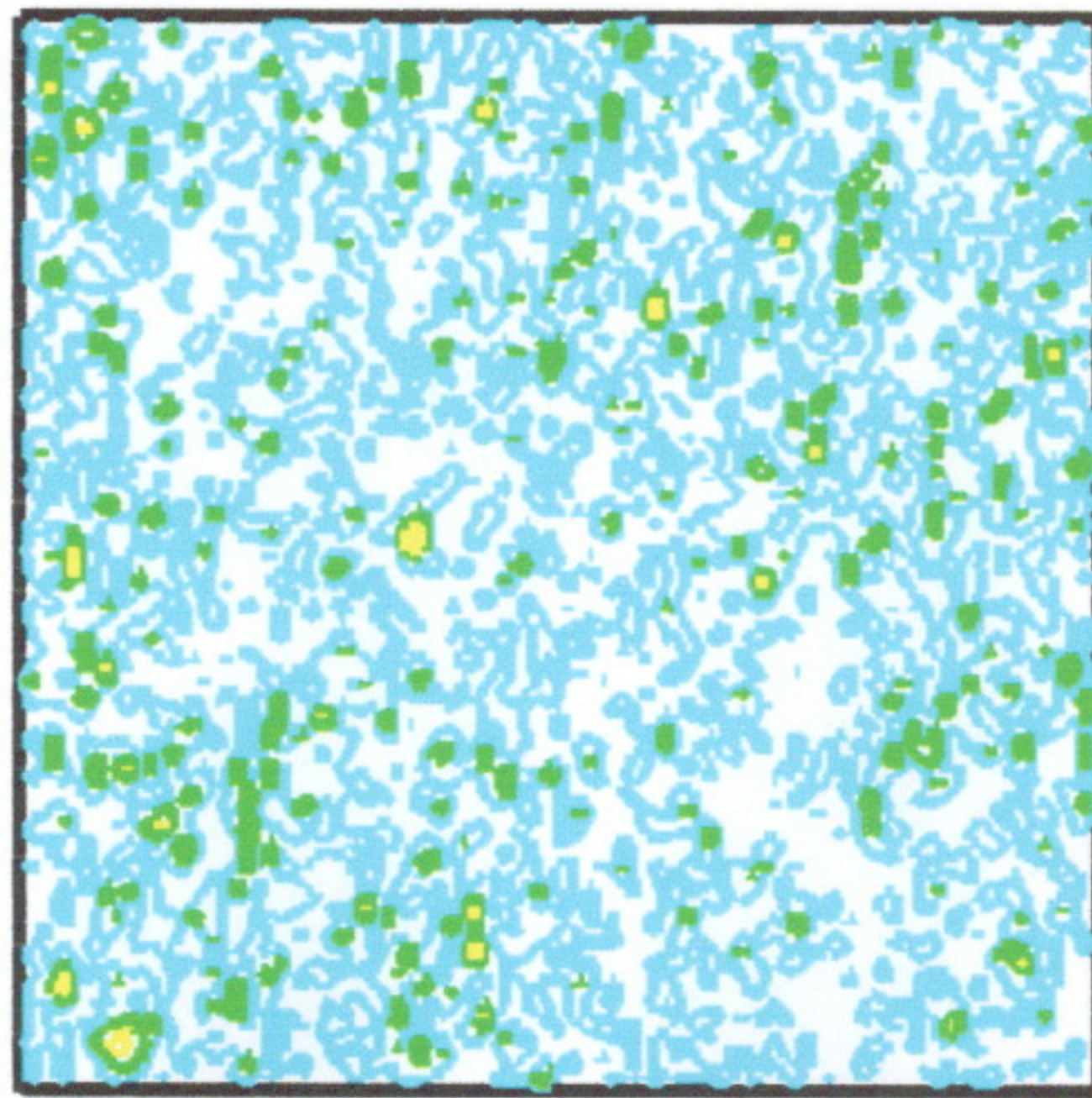WG Vincenti nd CH Kruger Jr 1965 *Introduction to Physical Gas Dynamics*, John Wiley & Sons, NY.

Figure 1. Contour for Magnitude of Velocity $|\boldsymbol{u}|$ at $t = 0.024$.
$n_{10} = 0.5$, $n_{20} = 0.25$, $\alpha_0 = 0.5$, $\rho_0 = 1.0$, $T_0 = T_w = 0.6$ (3,000 K),
$T_d = 11.9$, $\rho_d = 4 \times 10^7$, $K_n = 0.005$, $M_0 = 1.7$, $R_{e0} = 500$.
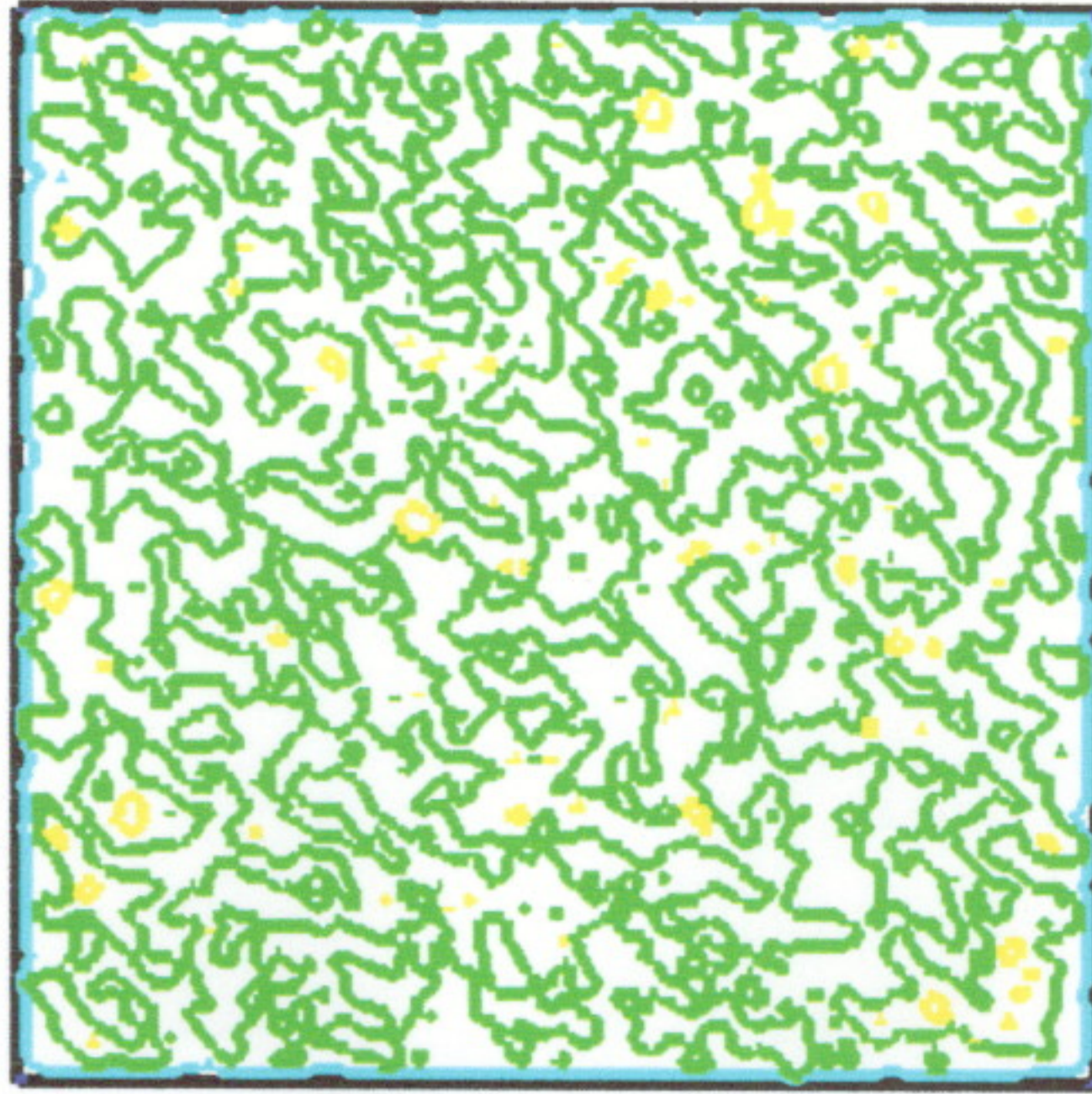
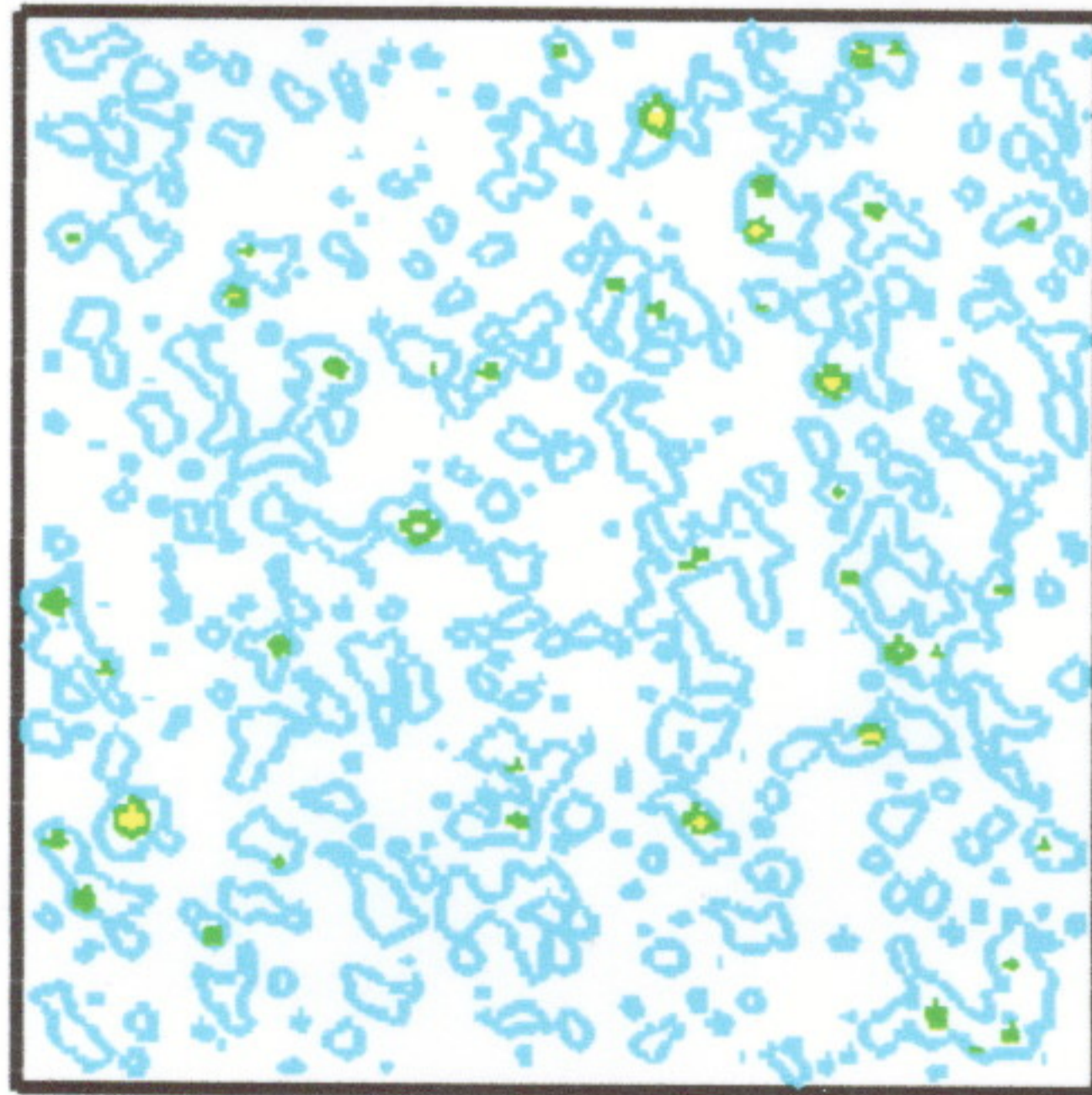Figure 2.   Contour for Temperature T at $t = 0.024$.

Figure 3.   Contour for Mass Fraction of Oxygen Atom α at $t = 0.024$.